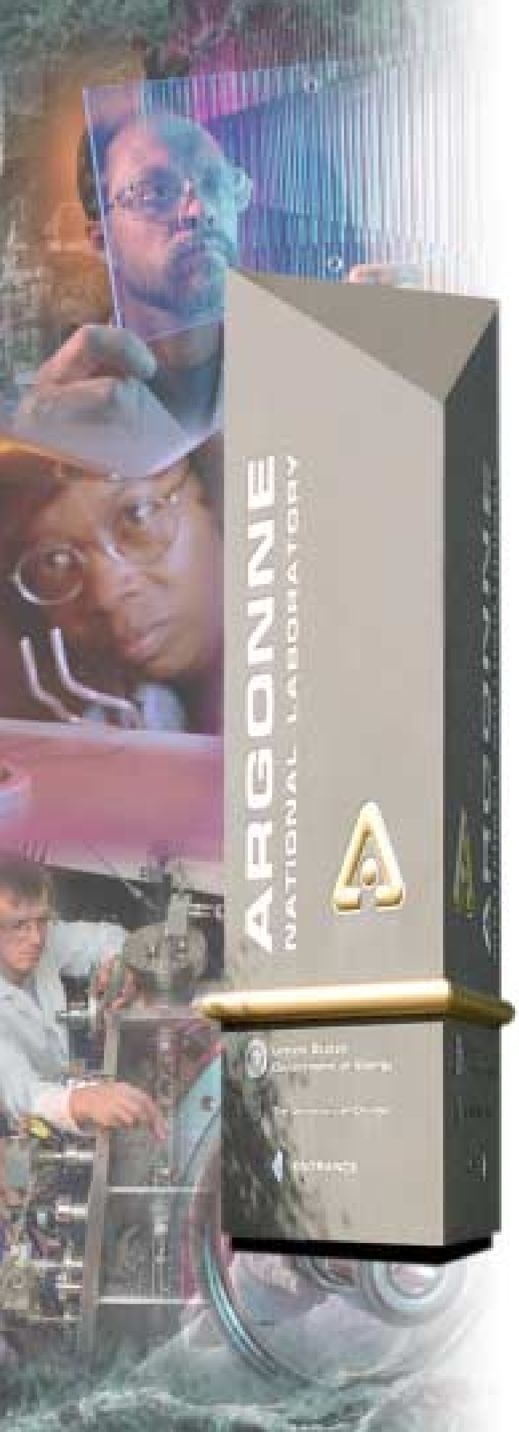

Overview of Advanced Alloy Post-Quench Ductility Program

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Review of ANL LOCA and Dry-Cask-Storage Programs
Argonne National Laboratory
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Argonne National Laboratory

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Advanced Alloy Post-Quench Ductility Scope

- **Program Purpose**
  - Using same apparatus, test conditions and data-analysis methods, determine post-quench embrittlement ECR for cladding alloys
  - Compare advanced alloy performance to Zry-4 and Zry-2
  - Determine why different Zr-1%Nb alloys behave so differently
- **Cladding Alloys**
  - M5, (F Zry-4), W ZIRLO, W Zry-4, (GNF Zry), "Fortum" E110
- **Approach**
  - 2-sided steam oxidation (1000-1260°C) to 5-20% CP-predicted ECR
  - Cool-down (≈10°C/s) to 800°C followed by water quench
  - ECR embrittlement threshold: ring compression & 3-point-bend tests
  - 4-point-bend of LOCA Integral Test Samples (balloon/burst/quench)

Description of Alloys

- **Alloys**
  - Zry-2 cladding (10×10, 10.3-mm OD, 0.66-mm wall, ≈10% Zr liner)
  - Zry-4 cladding (17×17)
    Westinghouse (W) (9.50-mm OD, 0.57-mm wall)
    Framatome (dimensions nominally the same as M5)
  - Framatome M5 cladding (17×17)
    Validation cladding (9.49-mm OD, 0.57-mm wall)
    "Data" cladding (9.50-mm OD, 0.61-mm wall)
  - W ZIRLO cladding (17×17, 9.50-OD, 0.57-mm wall)
  - TVEL E110 supplied by Fortum from Loviisa Plant in Finland
    Tubing (9.17-mm, 0.71-mm wall)
    Etched and anodized cladding (9.13-mm OD, 0.7-mm wall)
    Autoclave-oxidized cladding (to be provided)

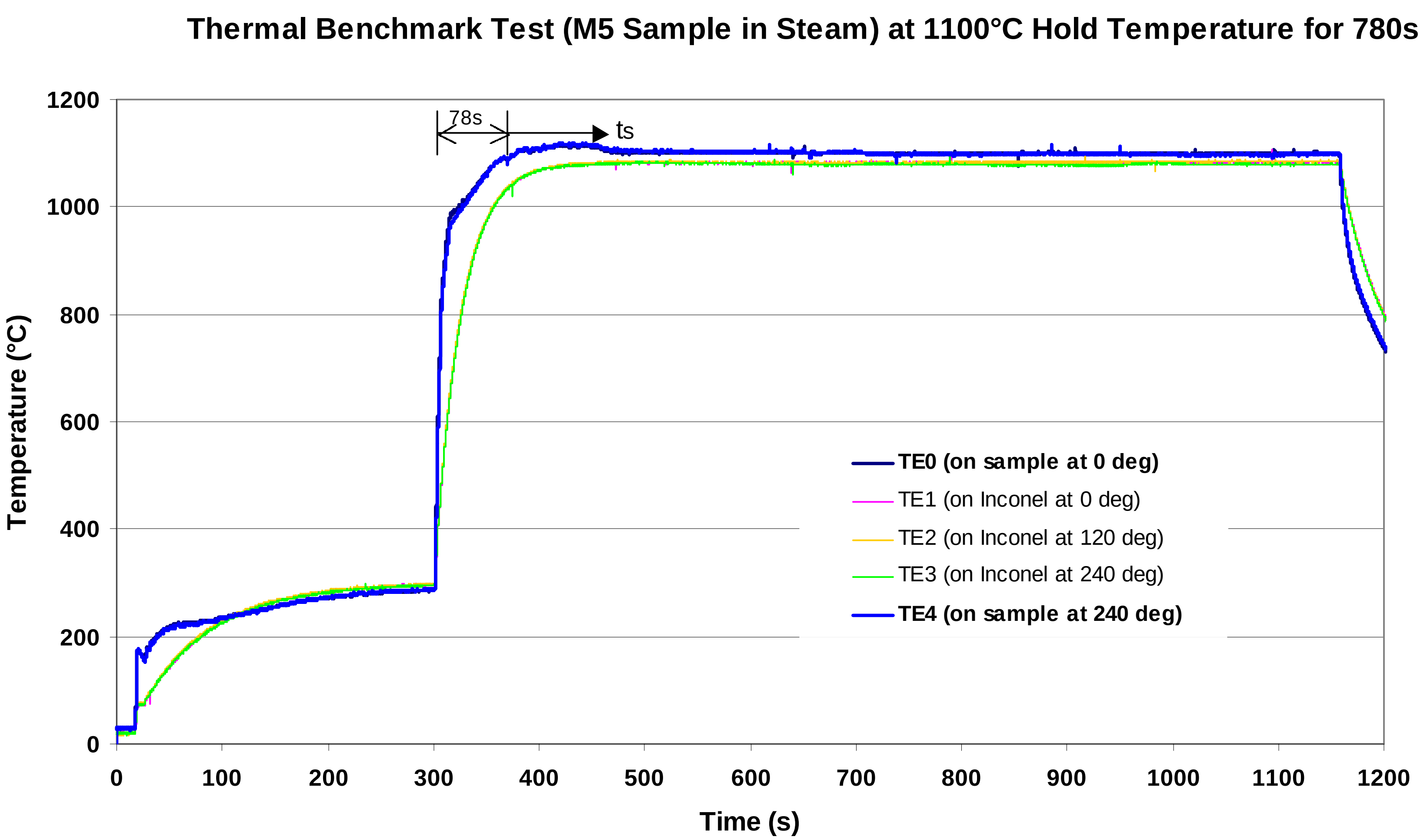
ECR and Heating Rate Considerations

- **Licensing basis (ECR ≤ 17%, T_{max} ≤ 1204°C)**
  - Well defined for T > 1050°C, assuming no breakaway oxidation
  - Temperature-time history may be more relevant for T < 1050°C
  - Test time depends on wall thickness and weight gain model (See Table for 2-sided oxidation test times to reach 17% ECR at 1000°C; test times for ballooned/burst region may be 40% less)
- **Heating Rate and Hold-Temperature Considerations**
  - LOCA-relevant: ≈0.5 to 10°C/s (poor for kinetics studies)
  - Fast-heating rates : ≈20 to 100°C/s (good for kinetics studies)
  - Compromise: fast-to-slow ramp to minimize T-overshoot (<20°C)
  - Hold temperature (1000, 1100, 1200°C) variation (<10°C average)
  - Oxidation behavior of some alloys is very sensitive to heating rate

2-Sided Test Times to Reach 17% ECR at 1000°C

Weight Gain Correlation	E110 (0.71 mm) Test Time, s	M5 (0.57 mm) Test Time, s
Baker-Just	3010	1940
Cathcart-Pawel	3770	2430
WG Constant based on ANL Polished E110 Data	6030	3890
WG Constant based on ANL M5 Data	10,880	4660
WG Constant based on CINOG Data for M5	24,330	15,680

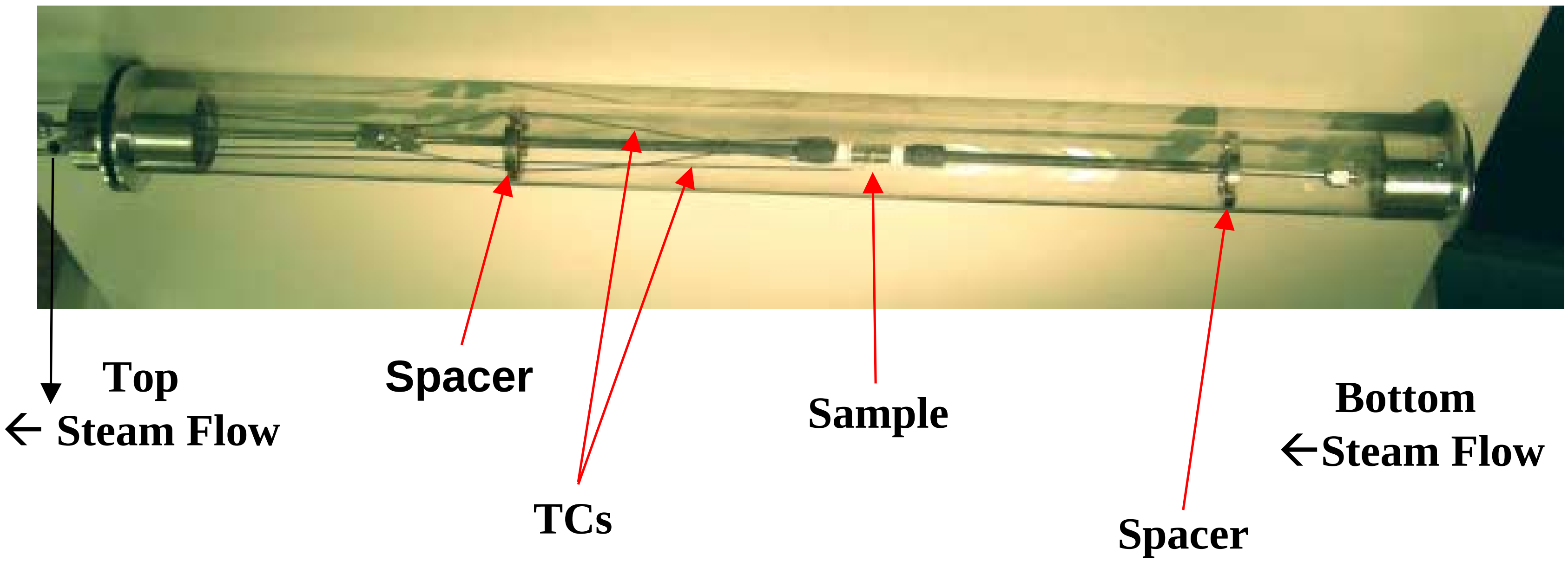
Temperature History for Controlled Ramp to 1100°C



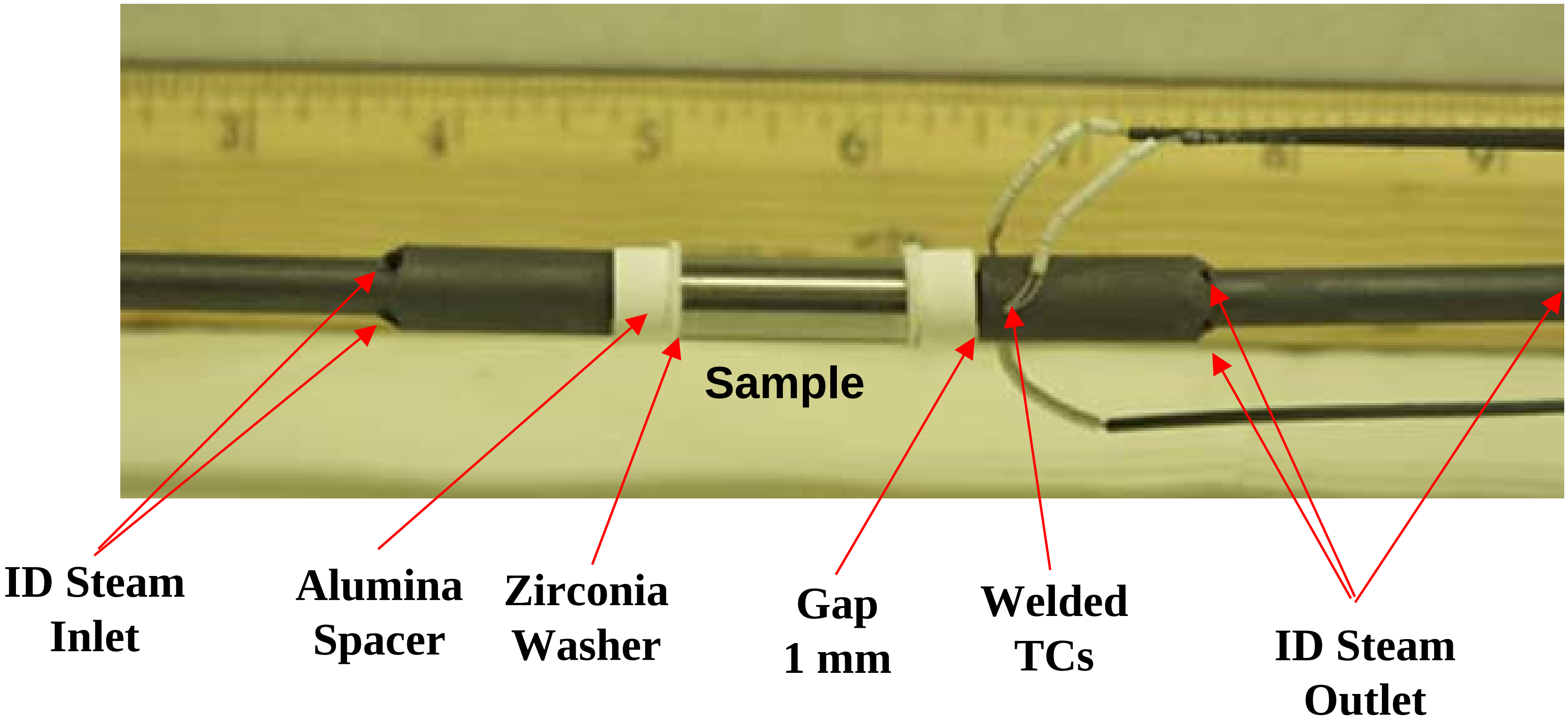
Heating Method and Temperature Monitoring

- **Heating Method**
  - Radiant heating deposited on 1-2 μm of outer surface
  - Focal area of furnace has diameter > cladding diameter
- **Furnace Power Controlled by TC on Inconel Holder**
  - 3 TCs (120° apart) on Inconel holder just above the sample
  - Capability to switch control TC
- **Thermal Benchmark Tests**
  - 2 TCs welded onto cladding sample outer surface
  - Correlate sample temperature history with control TC history
- **Why Tests are not Run with TCs Welded onto Sample**
  - Practical reasons: time consuming and expensive
  - Potential for disturbing oxide layer, especially for E110

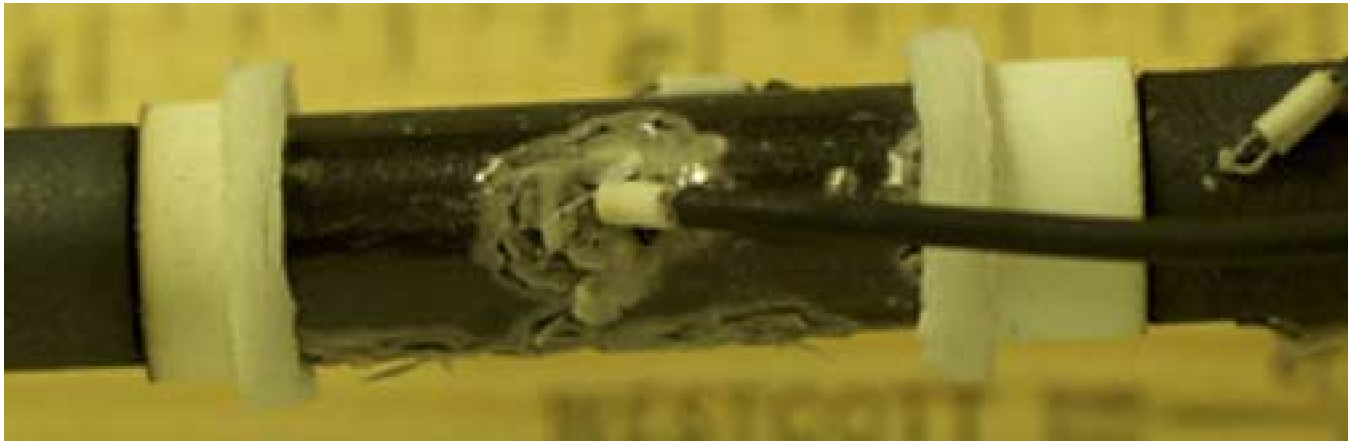
Steam Oxidation Test Train with Quartz Tube

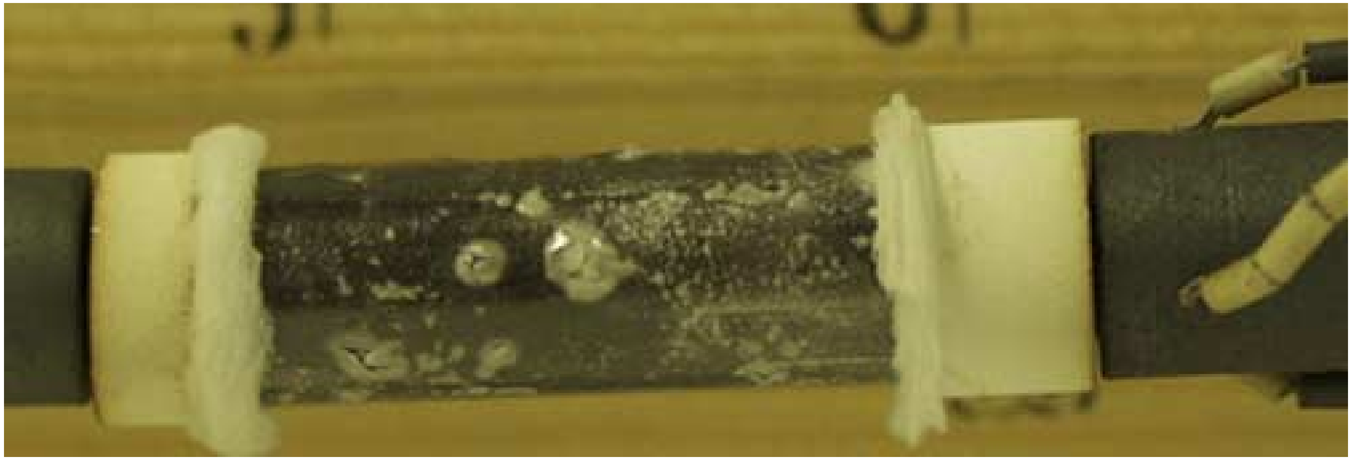


Two-Sided Steam Oxidation Test Train

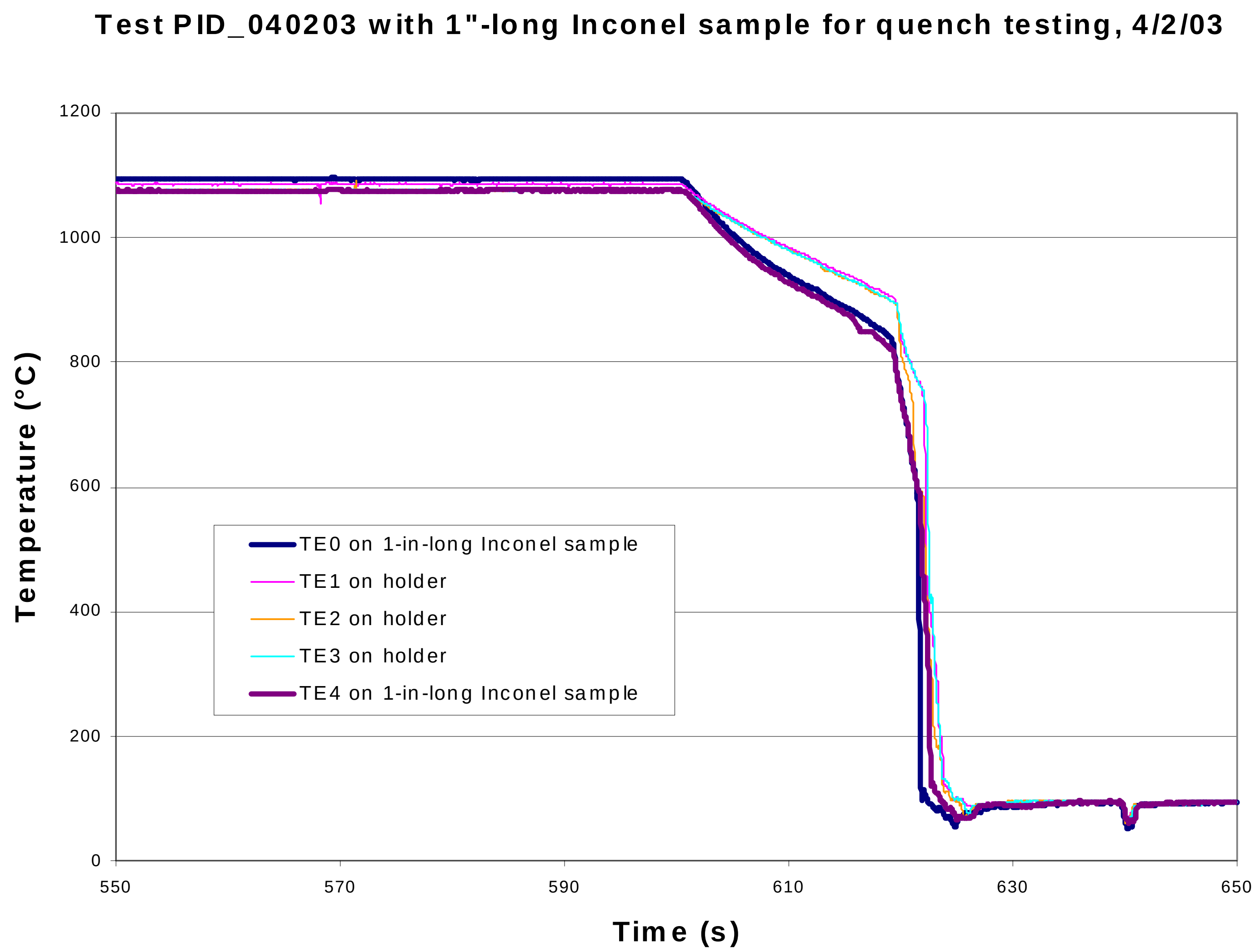


Effect of Welded TC on E110 after 290 s at 1000°C





Quench Sample Following Cool-Down to 800°C

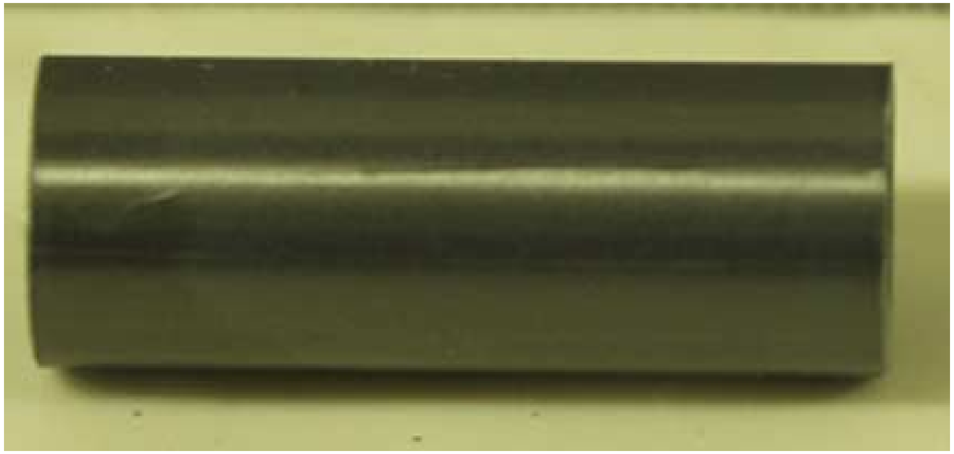


Embrittlement Mechanisms

- **Protective Oxide Layers (Lustrous Black, Tetragonal)**
  - Thinning of effective prior-beta layer with time, WG and ECR
  - Increase in oxygen content in prior-beta layer with increasing T
  - Effect of hydrogen from in-reactor corrosion; LOCA ballooning/burst
- **Classical Breakaway Oxidation for Zry-4 and M5**
  - Black (tetragonal)-to-white (monoclinic) transition on outer oxide surface
  - Increase in oxygen pickup rate; possibly hydrogen uptake
  - Generally not within LOCA-relevant times (e.g., after 3 h at 1000°C)
- **Nodular Breakaway Oxidation for E110**
  - Local enhancement of oxidation rate (e.g., E110 at 1100°C)
  - Local enhancement of hydrogen uptake (e.g., E110 at 1100°C)
  - Global enhancement of O and H uptake (e.g., E110 at 1000°C)

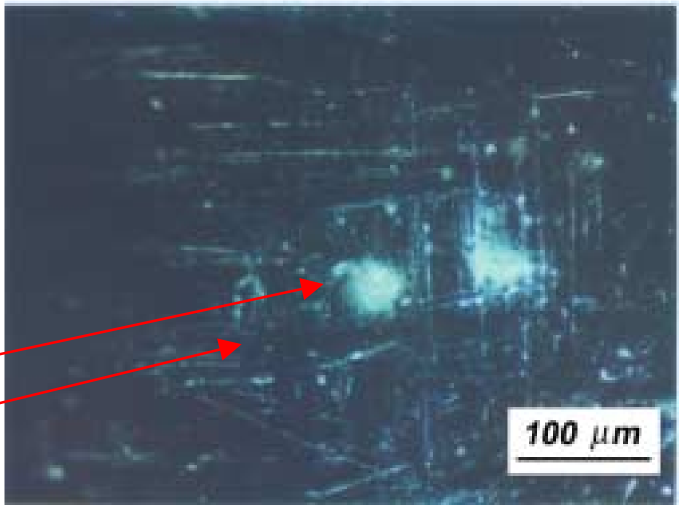
Protective Lustrous Black Oxide Layers

Zry-4
After 868 s (18% ECR)
in Steam at 1100°C



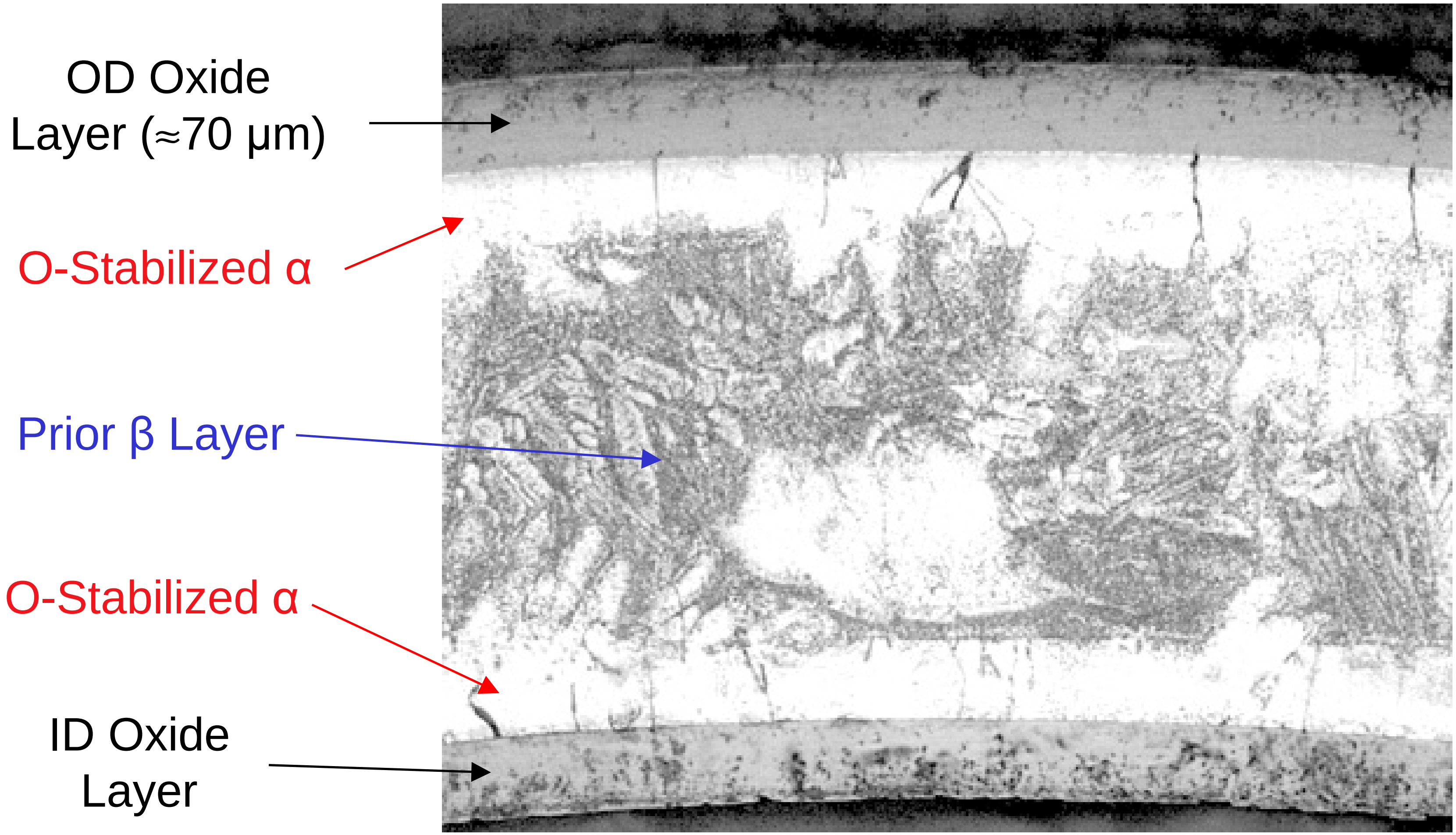
E110
After 75-s Ramp/5-s Hold
in Steam at 1000°C
(high magnification)

White Spots in
Lustrous Black Matrix

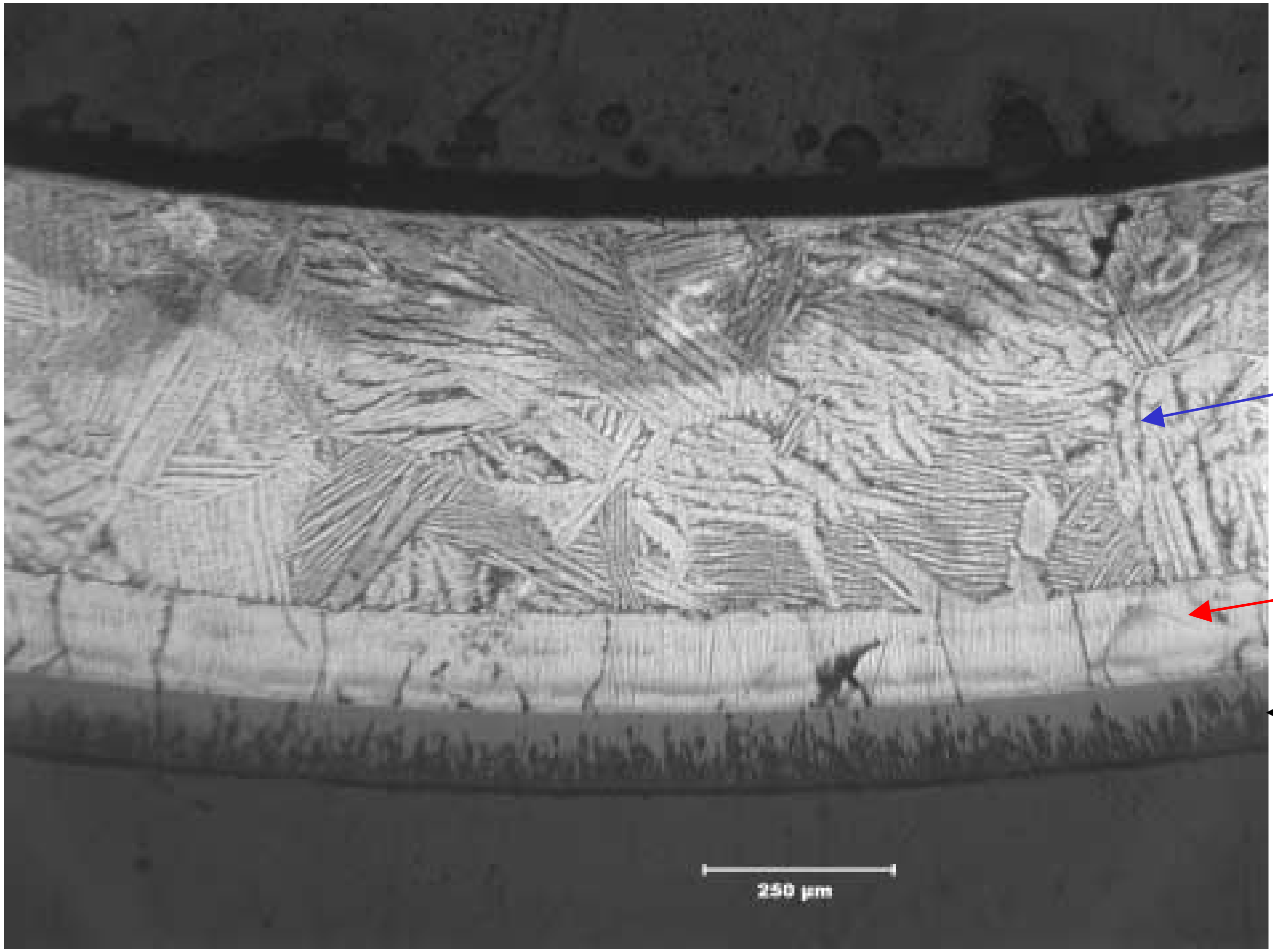


High Magnification of Protective Oxide Layer

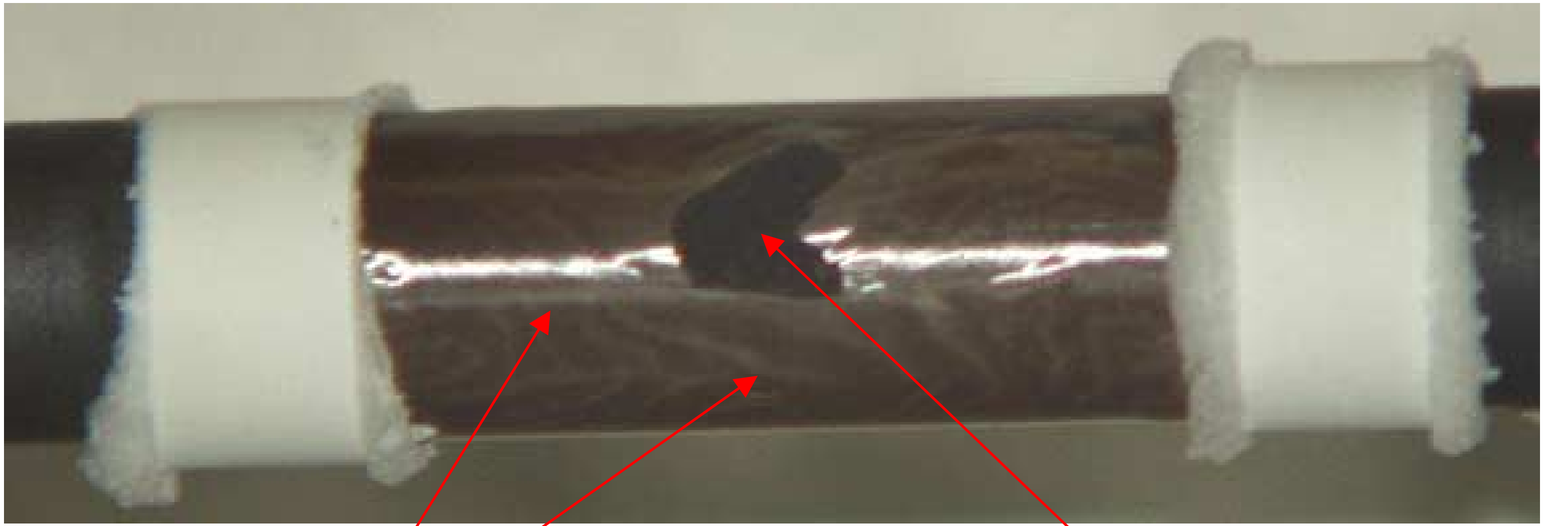
Zry-4, 1100°C for 868 s, 18% ECR, ΔH = 8 wppm



One-Sided Oxidation of Zry-2 at 1200°C for 10 Minutes



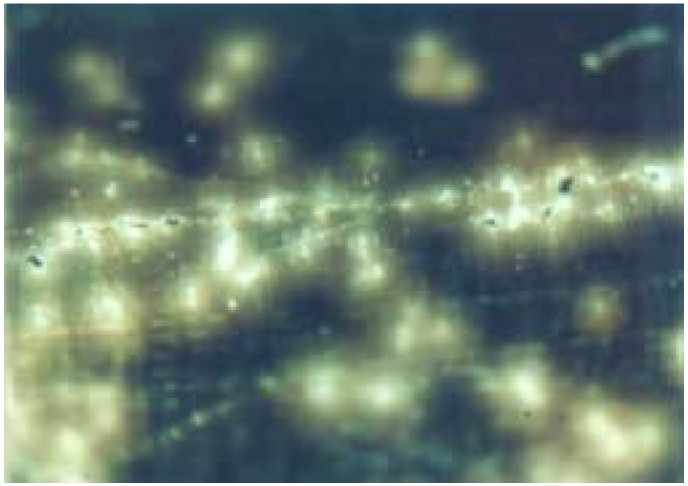
Classical Breakaway Oxidation: M5 after 3 h at 1000°C



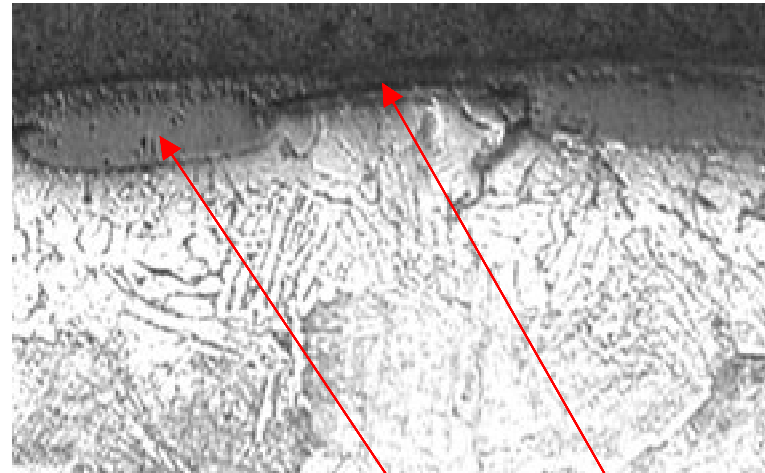
Mixed
Light-Dark
Layer

Dull Black Oxide
Under
Delamination

Nodular-to-Global Breakaway Oxidation in E110



1100°C
for
≈2500 s
(1-sided)



White Oxide

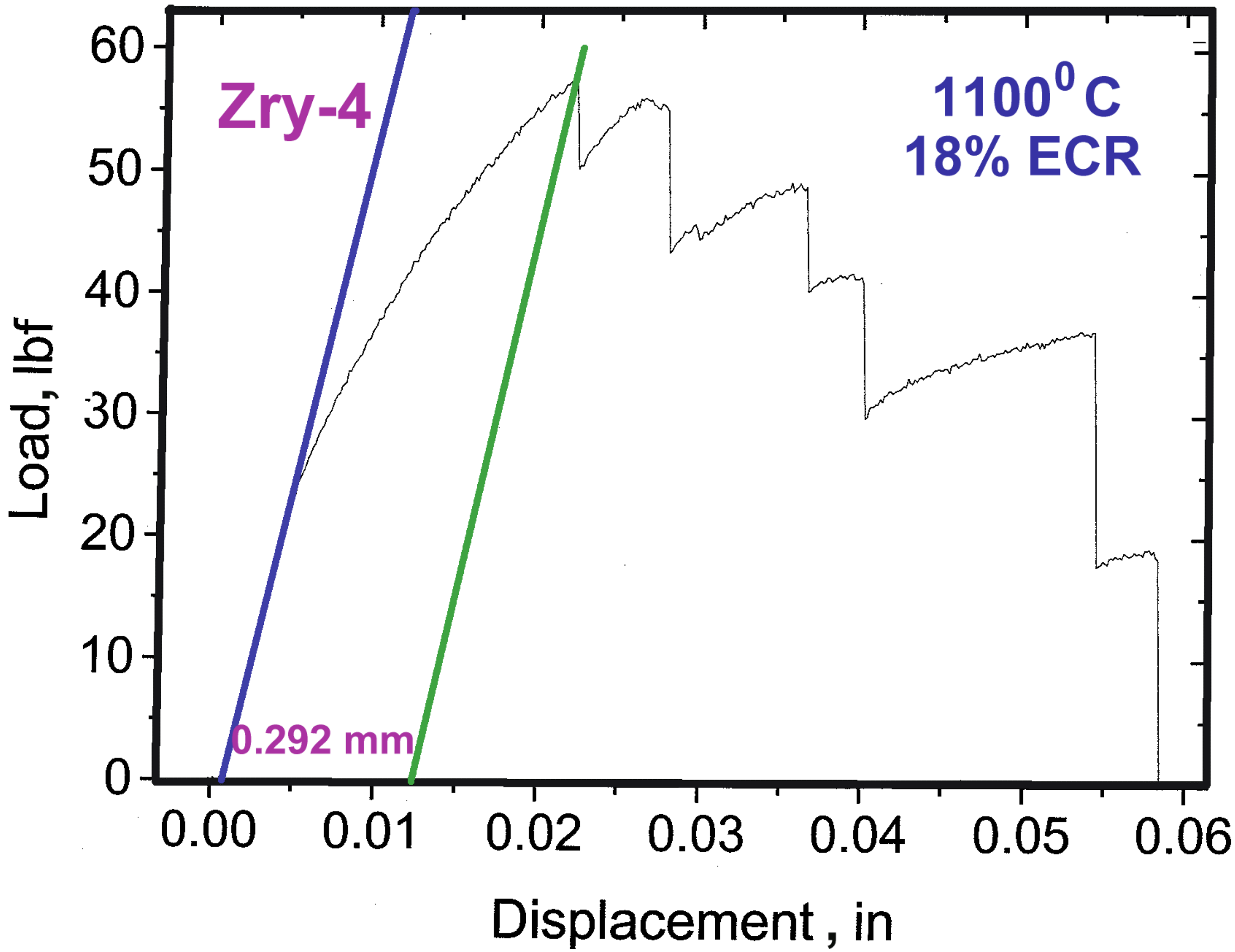
Black Oxide

1000°C
for 1400 s
(2-sided)

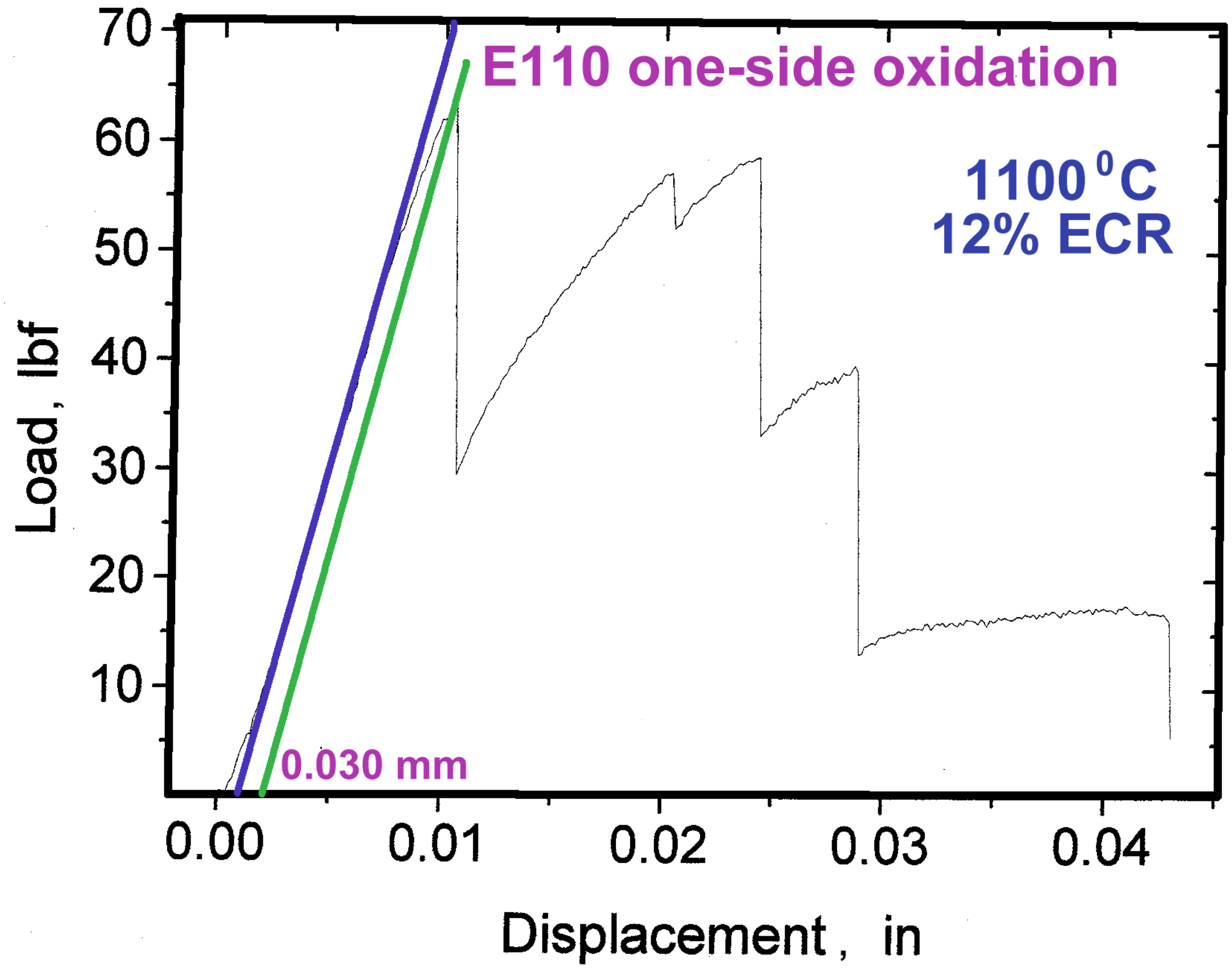
Post-Quench-Ductility Test Methods

- **Ring Compression Tests**
  - RT screening tests at 2 mm/min (0.35%/s) for 8-mm-long rings
  - Measure off-set displacement (δ_p) vs. ECR (5, 10, 17, 20% CP-model)
  - Convert to "nominal" strain ($\varepsilon = \delta_p/D_o$) vs. ECR
  - For alloys that embrittle at <17% ECR, repeat test at 135°C
- **3-Point-Bend Tests to Refine Transition ECR**
- **4-Point-Bend Tests following LOCA Integral Test**
  - See ECR and H distributions for Zry-2 after 1200°C for ECR_{max} = 17%
  - Burst region may act as a hinge by inducing failure for uniform M_b
  - Rings cut away from burst region may be brittle due to high H uptake
- **Testing of Pre-Hydrided Cladding (Phase 2) and High-Burnup Cladding (Phase 3) would Yield Valuable Data**

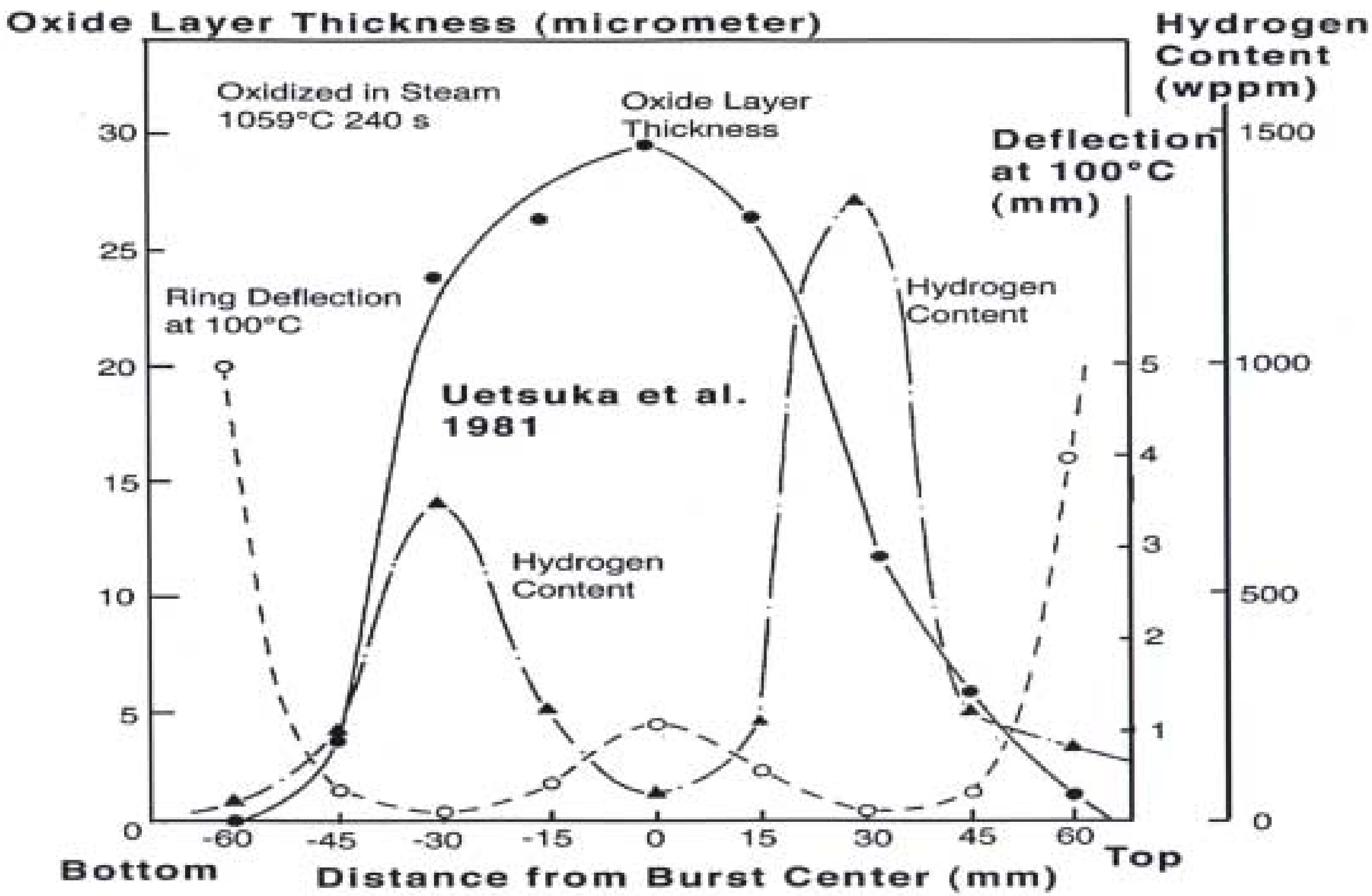
Zry-4 Ring-Compression Results after 18%ECR at 1100°C



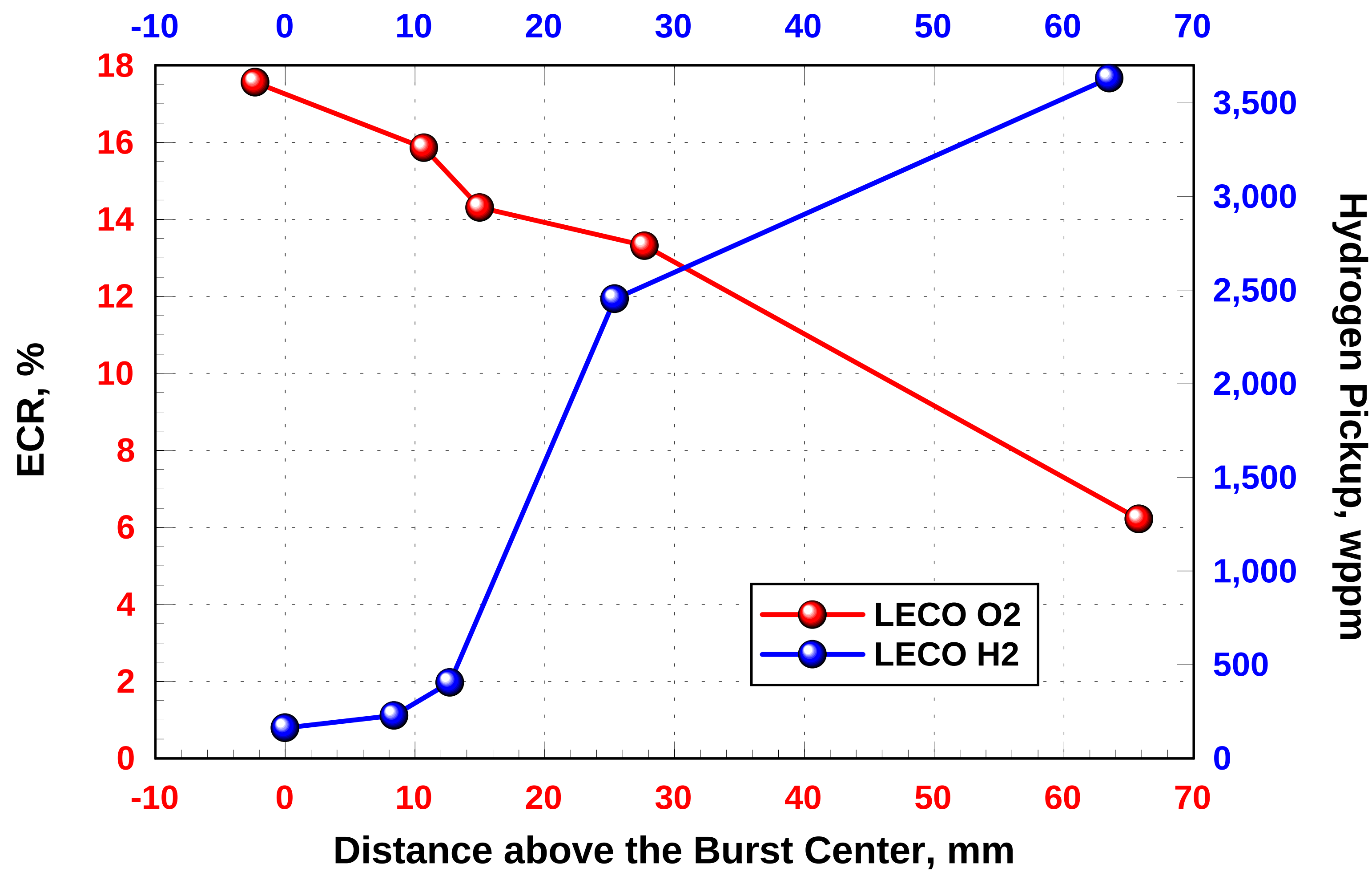
E110 Ring-Compression Results after 12%ECR at 1100°C



JAERI LOCA Integral Test Results (1981)



LOCA Integral Test Results for Zry-2: 1200°C for 5 Min.



Summary of Approach to Post-Quench Ductility Tests

- **Extensive Verification/Validation Studies without Quench**
- **Limited Verification/Validation Studies with Quench**
- **Oxidize Alloys for Same Test Times to ≤20% ECR (0.57-mm Wall and Cathcart-Pawel WG Model) and Quench**
  - ≤3400 s (1000°C), ≤1100 s (1100°C), ≤400 s (1200°C), ≤230 s (1260°C)
- **Determine "Measured" ECR Based on Weight Gain**
- **Oxidation Kinetics and Post-Quench Ductility Data**
  - Compare results for ZIRLO and M5 to Zry-4 (and Zry-2) data
- **Explore Factors that may Contribute to E110 Behavior**
  - Confirm poor post-quench ductility performance at low test times (ECRs)
  - Explore effects of changing surface roughness and chemistry
  - Characterize: bulk chemistry, metallography, SEM, TEM